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FINAL REPORT

for

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UT/NASA Multidisciplinary Research Grant

on

High Resolution Raman and Infrared Spectra of  $\text{NO}_2$  and  $\text{SO}_2$

by

Professor William H. Fletcher, Research Director in Chemistry

and

Associate Professor Kenneth Fox, Research Director in Physics and Astronomy

for period

21 June 1971 to 31 December 1972

Personnel

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Research Associates: Dr. J. S. Rayside (Chemistry  
Dr. G. D. T. Tejwani (Physics & Astronomy)

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& Astronomy

(NASA-CR-138420) HIGH RESOLUTION RAMAN  
AN INFRARED SPECTRA OF  $\text{NO}_2$  AND  $\text{SO}_2$   
Final Report, 21 Jun. 1971 - 31 Dec.  
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## I. Infrared Spectrum of Sulfur Dioxide

The infrared-active vibration-rotation fundamentals of sulfur dioxide,  $^{32}\text{S}^{16}\text{O}_2$ , have been measured with moderately high spectral resolution. Quantum number assignments have been made for spectral lines by comparison with theoretically computed line positions and intensities, which include the effects of centrifugal distortion. New improved values for the band centers have been determined. Intensities of the observed lines have also been computed. Dipole moment derivatives have been obtained. The technical details of this work were reported<sup>1</sup> at the Ohio State University Symposium on Molecular Structure and Spectroscopy, Columbus, Ohio held during 12-16 June 1972; in papers published in the Journal of Chemical Physics<sup>2</sup> and in Chemical Physics Letters<sup>3</sup>; and in a research report<sup>4</sup> comprising important data too extensive to be published in the open literature.

In addition, the infrared-active vibration-rotation combination band  $\nu_1 + \nu_3$  of  $^{32}\text{S}^{16}\text{O}_2$  has been measured with moderately high spectral resolution. Quantum number identifications of spectral lines were made by comparison with theoretically computed spectra which include the effects of centrifugal distortion. Relative line intensities were also calculated. A new improved band center for  $\nu_1 + \nu_3$  has been determined. The technical details of this work were presented extensively in a research report<sup>5</sup>. It is anticipated that short summaries will also be given at an open meeting and in the open literature in 1973. Some implications of this work for air pollution were discussed<sup>6</sup> at the Symposium on the Use of Astronomical Techniques for the Study of Atmospheric Deterioration, held at the University of Washington, Seattle, Washington on 13 April 1972.

Self-broadened and foreign-gas ( $N_2$  and  $O_2$ ) pressure-broadened linewidths of  $SO_2$  and  $NO_2$  have been calculated. Computed values of linewidths are in good agreement with all the available experimental results and other reported theoretical calculations on four self-broadened and one nitrogen-broadened line based upon another theory. Air-broadened linewidths have been calculated for  $SO_2$  at 200, 250, and 300°K. The technical details of this work were reported<sup>7</sup> at the First International Conference on Spectral Lines, held at The University of Tennessee, Knoxville, Tennessee during 28 August - 1 September 1972; in a paper<sup>8</sup> published in the Journal of Chemical Physics; and in a research report<sup>9</sup> comprising important data too extensive to be published in the open literature. Additional calculations related to self-broadened pure rotation transitions recently measured in  $SO_2$  have been submitted for publication<sup>10</sup>.

The measurement and analysis of the infrared-active vibration-rotation fundamental bands of nitrogen dioxide were not completed primarily for two reasons. First, technical difficulties related to the need for measurements at high temperatures proved insurmountable with our limited equipment. And second, it was brought to our attention privately that data of higher resolution had been obtained elsewhere, and was being analyzed. Consequently, we judged that our time and resources could be better spent in extending our work on sulfur dioxide. Should suitable resources become available to us in the future, we would reconsider a program of measurements and analyses of  $NO_2$ .

Our results are potentially applicable to studies of the terrestrial atmosphere where sulfur dioxide and nitrogen dioxide play serious roles as

pollutants. High-resolution infrared spectroscopy is a possible technique for the remote detection and monitoring of these molecules in situ. For example, monochromatic laser emissions may be useful for studying terrestrial  $\text{SO}_2$  in absorption. We found several relatively isolated and moderately strong  $\text{SO}_2$  lines in our laboratory spectra, with observed positions which fall close to observed laser oscillations.<sup>1,2,4</sup> Although the spectral coincidences are not exact for all of the lines, nevertheless the spectral broadening by air will produce sufficient overlapping of the lines in question.<sup>7-9</sup> It is also possible that the  $\nu_1 + \nu_3$  band of terrestrial  $\text{SO}_2$  may appear in solar spectra, and be susceptible to analysis there. Our intensity studies will aid in the determination of concentrations of atmospheric  $\text{SO}_2$ .

Incidentally, we note that the  $\nu_1 + \nu_3$  band of  $\text{SO}_2$  was recently considered in a determination of detectability of this gas in the atmosphere of the planet Mars. An upper limit approximately 300 times smaller than the terrestrial atmospheric value was established, on the basis of the  $\nu_3$  fundamental, for  $\text{SO}_2$  in the Martian atmosphere.

There is considerable value, from a fundamental spectroscopic viewpoint, in studying gases like  $\text{SO}_2$  and  $\text{NO}_2$  at even higher resolution in the infrared. Such studies would permit a more complete determination of vibration-rotation line positions and intensities, as well as quantum-number assignments. Further, it would be possible to measure line-shapes and line-broadening parameters as a function of vibration and/or rotation quantum numbers.

## II. Raman Spectroscopy

Our original goal was to measure the fundamental vibration-rotation Raman bands of  $\text{SO}_2$  and  $\text{NO}_2$  in the gas phase with a resolution in the range

of  $0.1 \text{ cm}^{-1}$ , and to analyse the observed spectra in a manner similar to that which was done for the infrared bands. The spectrometer which was to be used for measuring the Raman spectra was constructed in the Chemistry Department, and it had barely been completed at the beginning of this project. Our initial work with  $\text{SO}_2$  showed that the spectrometer would indeed produce the resolution expected, but some improvements would be required in order to produce spectra of the best quality. The following major improvements were made:

1. A new and improved optical system was constructed for focusing and multi-passing the laser beam through a gas sample. It also serves to collect the Raman radiation and focus it onto the monochromator slit.
2. The air conditioning and filtering system was replaced and improved in order to reduce the concentration of dust particles in the room. (The cost of this change was borne by the University.)
3. The photon counting system was modified to reduce the level of random noise in the recorded signal. It is now functioning very near the lowest possible noise level.

It was necessary to send our photomultiplier housing and refrigeration system back to the manufacturer for some of these modifications. At the writing of this report the equipment has been back in our hands only four days, and this has permitted us time only for some scans of the  $\nu_1$  band. This band is not as strong as we had hoped to find it. It appears that the intensity in the P and R branches of the band is such that  $0.2 \text{ cm}^{-1}$  is the minimum slit width that we can use for observations of  $\nu_1$  and  $\nu_3$ . This should be sufficient to perform an analysis of at least  $\nu_3$  by computing the expected spectrum in the same manner as was done for the fundamentals in the infrared spectrum.

The Q branch of  $\nu_1$  is about 500 times more intense at the peak than the maximum intensities in the P and R branches. The Q branch was scanned with a spectral slit width slightly less than  $0.10 \text{ cm}^{-1}$ , and its maximum was observed at  $1151.95 \text{ cm}^{-1}$ . This maximum is not expected to coincide with the band origin, so this value is in satisfactory agreement with the value of  $1151.65 \text{ cm}^{-1}$  determined from the infrared spectrum.<sup>4</sup>

It should be noted here that, while we have not completed the work originally proposed because of the necessity of improving the spectrometer system, the work of scanning and analyzing the fundamentals of  $\text{SO}_2$  and  $\text{NO}_2$  will proceed under alternative funding, and the results will be published in the open literature.

Our preliminary theoretical studies relevant to Raman spectra of  $\text{SO}_2$  and  $\text{NO}_2$  indicate a high probability of success in analyzing high-resolution laser Raman spectra of gases. These studies have led us to generate a new proposal, submitted to the Office of Naval Research (Project Squid), on the measurement and analysis of line positions, intensities, and pressure-broadened widths in vibration-rotation Raman bands of gases. This proposal has received a favorable preliminary review, on the basis of which we have been invited to make an in-person presentation of our research plans.

### III. Impact of Grant on The University of Tennessee

The impact of this grant on The University of Tennessee, Knoxville, has been significant. One effect is the establishment of the Earth Resources and Astrophysics Laboratory (ERAL) in the Department of Physics and Astronomy, with Dr. Kenneth Fox as its director. Currently, research programs in spectroscopy of atmospheres, supported by the National Science Foundation and the Air Force Cambridge Research Laboratories, are being carried out in this Laboratory under the direction of Dr. Fox.

Another effect has been the infusion of new personnel into the research programs of the University and the Laboratory. In particular, the UT/NASA grant made it possible to attract Dr. Tejwani to the University. He recently received his Ph.D. from the State University of New York at Stony Brook, and is already recognized internationally for his work on pressure-broadening calculations in spectroscopy. We have been able to support Dr. Tejwani beyond the termination of the UT/NASA grant, by means of other programs in ERAL. During the Summer of 1972, Dr. Bernard Bobin from the Laboratory of Molecular Spectronomy, University of Dijon, France, was a postdoctoral research associate in our Laboratory. Also, currently doing research in ERAL are Ms. Francoise Michelot (from Dijon) and Mr. Raymond J. Corice, Jr.

This grant has also made it possible to complete the construction of a high resolution Raman spectrometer system for gases and to establish a Raman Spectroscopy Laboratory, which is under the direction of Dr. Fletcher in the Department of Chemistry. It has also provided funds to hire Dr. John S. Rayside, who has had prior experience in laser Raman spectroscopy and with photon counting systems. Dr. Rayside will continue work in

this laboratory under funding from NSF. It is probable that the work in progress under the support of this UT/NASA grant was a significant factor in securing the grant from NSF.

Effects related to national interests in extending knowledge in the fields of aeronautics and space may be illustrated with some specific examples. Numerous requests for preprints and reprints of our work, as well as suggestions for future research bearing on national interests, have come from universities such as the University of Chicago and the University of Washington; from government agencies, such as the Environmental Science Services Administration; and from industries such as the Honeywell Radiation Center. In particular, copies of two letters, one from the Jet Propulsion Laboratory (which is supported primarily by NASA) and the other from the U.S. Army Ballistic Research Laboratories, concerning the relevance our work are attached in Appendix I. A general awareness of our research in ERAL is reflected in a letter to Science (copy in Appendix I) describing some of our activities.

Appendix I. Copies of Letters Related to the Impact of the UT/NASA Grant

Appendix II. Copies of Published Papers and Reports (15 of each)



# References

1. "Experimental and Theoretical Studies of the Fundamental Bands of SO<sub>2</sub>," R. J. Corice, K. Fox, and G. D. T. Tejwani, Proceedings of the Ohio State University Symposium on Molecular Structure and Spectroscopy, Columbus, Ohio, 12-16 June 1972, Abstract W2.
2. "Experimental and theoretical studies of the fundamental bands of sulfur dioxide," R. J. Corice, K. Fox, and G. D. T. Tejwani, Journal of Chemical Physics, **58**, 265 (1973).
3. "Dipole Moment Derivatives for SO<sub>2</sub>," G. D. T. Tejwani, K. Fox, and R. J. Corice, Chemical Physics Letters \_\_\_\_\_, (1973).
4. "Fundamental Bands of <sup>32</sup>S<sup>16</sup>O<sub>2</sub>," K. Fox, G. D. T. Tejwani, and R. J. Corice, Research Report No. UTPA-ERAL-01, September 1972.
5. "ν<sub>1</sub> + ν<sub>3</sub> Combination Band of SO<sub>2</sub>," R. J. Corice, K. Fox, and G. D. T. Tejwani, Research Report No. UTPA-ERAL-03, December 1972.
6. "Intensity Studies and the Surveillance of Molecules in the Terrestrial Atmosphere," K. Fox, D. N. B. Hall, and W. S. Benedict, Proceedings of the Symposium on the Use of Astronomical Techniques for the Study of Atmospheric Deterioration, University of Washington, Seattle, Washington, 13 April 1972, Abstract 1.
7. "Calculation of Pressure Broadened Line Widths of SO<sub>2</sub>," G. D. T. Tejwani, Proceedings of the First International Conference on Spectral Lines, The University of Tennessee, Knoxville, Tennessee, 28 August - 1 September 1972, Abstract 1C1.2.
8. "Calculation of Pressure-Broadened Linewidths of SO<sub>2</sub> and NO<sub>2</sub>," G. D. T. Tejwani, Journal of Chemical Physics **57**, 4676, (1972).
9. "Computed Linewidths of SO<sub>2</sub>," G. D. T. Tejwani, Research Report. No. UTPA-ERAL-02, November 1972.
10. "Line Width Parameters for ΔJ = 1, 0 ≤ J ≤ 43 Rotational Transitions of the SO<sub>2</sub> Molecule", W. H. Yang, J. H. Roberts, and G. D. T. Tejwani, Submitted for publication.



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August 10, 1972

Refer to: 825-RAT:da

Professor Kenneth Fox  
Department of Physics and Astronomy  
University of Tennessee  
Knoxville, Tennessee 37916

Dear Ken:

Thank you very much for the preprint on  $\text{SO}_2$ . As you suggest, I will not circulate or distribute the manuscript. The contents are very interesting and I feel that your numbers for the  $\nu_3$  band will be quite useful to us. However, if I find it necessary to quote your numbers for any work (such as data reduction of flight data, etc.), I will contact you first.

Sincerely yours,

Robert Toth



DEPARTMENT OF THE ARMY Dr. Snider/ilm/301-278-  
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AMXBR-CA

19 Dec 72

Dr. Kenneth Fox  
Department of Physics & Astronomy  
University of Tennessee  
Knoxville, TN 37916

Dear Dr. Fox:

I want to thank you for sending me the reprints and reports covering your work on  $\text{SO}_2$ ,  $\text{NO}_2$ , and  $\text{CH}_4$ .

As you might know, we are currently using the technique of infrared emission spectroscopy to make minor constituent measurements in the upper atmosphere. Since the accuracy of this technique depends on the half-widths as well as the positions and intensities of the lines, your work is of considerable interest to us.

We would like to encourage you to continue this work, possibly extending it to other molecules of atmospheric interest, such as  $\text{O}_3$  and  $\text{HNO}_3$ .

Sincerely yours,

DONALD E. SNIDER  
Aeronomy Branch  
Concepts Analysis Laboratory

## Astronomy and Air Pollution

In May 1972, astronomer Charles G. Abbot of the Smithsonian Institution celebrated his 100th birthday in Washington, D.C. Several weeks before this event, astronomers gathered following a meeting of the American Astronomical Society in Seattle, Washington, to discuss progress in a field for which Abbot laid the foundation. The University of Washington sponsored a 1-day seminar entitled "The Use of Astronomical Techniques for the Study of Atmospheric Deterioration," at which the utilization of astronomical measurements to determine properties of the atmosphere and to monitor air pollution was discussed (1). Two topics were considered: the use of photometry to study the extinction of light by small particles in the atmosphere, and the use of spectroscopic techniques for determining the concentrations of molecular contaminants.

A comprehensive study of the aerosol content of the atmosphere in North America, South America, and Africa was provided by the efforts of C. G. Abbot and his co-workers, who determined the atmospheric extinction in connection with the Smithsonian's study of the solar constant from 1902 to 1950. Abbot was exceedingly careful to determine the transparency of the atmosphere accurately and frequently during the many years of the solar constant program. The extinction over the spectral range from 3500 Å to 1.6  $\mu\text{m}$  showed that the atmospheric aerosol content is lowest in the winter and highest in the summer, an effect also noted by others. A further correlation was found between the atmospheric transparency at all wavelengths and the humidity, in the sense that under circumstances of high humidity, the transparency is low. Scientists from the National Aeronautics and Space Administration (NASA) and the San Diego State College, analyzing Abbot's data, found a connection on a worldwide scale between the atmospheric extinction and volcanic activity. This connection was also noted by workers at Lowell Observatory in Arizona. In the latter case, however, the extinction measurements were carried out at night by utilizing photometry of the outer planets instead of direct photometry of the sun. At Lowell Observatory, at least, the average extinction for 1972 is not significantly different from the lowest value observed during the

1950's, which suggests that extinction variations in northern Arizona, until now, have been mainly affected by volcanic rather than human activity.

A large number of observatories around the world have submitted extinction data for possible atmospheric use to Project ASTRA (Astronomical and Space Techniques for Research on the Atmosphere) at the University of Washington. They have been examined sufficiently to show the presence of seasonal variations of extinction similar to those reported by NASA. There is also a definite decrease of extinction with altitude, which indicates an aerosol scale height of about 2 km. It is interesting to compare the data from two observatories in India, one in Hyderabad and one 56 km away from the city. The latter shows very large extinction values that are virtually indistinguishable from those obtained at the observatory in the city itself, which suggests a widespread uniformity of aerosol. Data collected from astronomers who have used Mt. Wilson have shown a steady deterioration over the last 60 years that can only be due to the increased pollution of the atmosphere in the Los Angeles valley. It was noted at the meeting that Mt. Wilson was abandoned by the Smithsonian as early as 1920 because of atmospheric pollution. Abbot had written (2), "Mount Wilson atmospheric conditions had proved unsuitable, and seemed steadily deteriorating for these exacting purposes as Los Angeles and neighboring towns expanded."

Among the surprising results obtained in the studies of atmospheric extinction was the discovery by several astronomers of what appeared to be an anomalous wavelength dependence. The extinction due to aerosol is greater in the red than in the blue. This seems to be an effect that occurs frequently only in sites where the aerosol content is extremely low. However, on the rare occasions when this anomalous wavelength dependence is in effect during periods of high aerosol content, a remarkable result occurs: Objects are blued instead of reddened, and a true "blue moon" can occur. Calculations by Project ASTRA scientists using the Mie equations suggest that for this to happen there must be a dominance of particles of radius near 0.5  $\mu\text{m}$ , the geometric standard deviations must be small, and the imaginary part of the refractive index cannot be larger than

about 0.01. Thus, the anomalous extinction characteristic itself may be a property of atmosphere aerosol that is subject to pollution effects by increases in light absorption by soot and similar particles.

Comparisons of nephelometric data with astronomical extinction measurements have provided new possibilities for the interpretation of each type of measurement in terms of the characteristics of particles and their atmospheric distribution. Reviews of problems in the interpretation of aerosol densities as determined by collections on filters, by lidar backscattering measurements, and by other techniques related the astronomical measurements more closely to traditional determinations of the nature and amount of atmospheric contaminants.

High-resolution spectroscopy has long been an important tool for investigating stellar and planetary atmospheres, and many of its techniques have been applied to the study of the minor constituents of the earth's atmosphere. The application of spectroscopic techniques in monitoring these constituents and their concentrations was discussed. In particular, it was suggested that certain spectral lines of  $\text{CH}_4$  might be used to monitor its terrestrial abundance and to serve as an atmospheric thermometer through its rotational temperature.

Researchers at the University of Tennessee, Kitt Peak National Observatory, the University of Hawaii, Pomona College, and the University of Washington have been studying several particularly noxious gases, such as  $\text{SO}_2$  and  $\text{NO}_2$ , in the laboratory and in the atmosphere, as well as theoretically. One novel approach involves a photometric system for matching the absorption minima and maxima of the spectrum of a pollutant in a restricted wavelength region with Fabry-Perot filters. The technique is suitable for use on small telescopes, and would thus allow scientists to measure the temporal variations of the contaminants in areas of moderate to heavy pollution by using either fixed urban telescopes or portable telescopes. It was suggested that it might even be possible to measure the movement of clouds of smog over the telescope. This technique has the advantage, too, that the spectrum of the star used as a source is not critical, as long as it is not variable and there are not too many deep absorption lines in the band under study.

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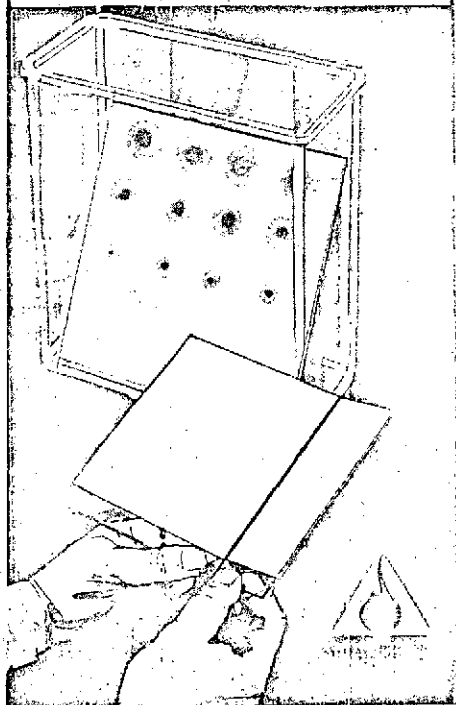
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Seattle meeting confirmed an opinion that is held by certain astronomers that their data are a useful adjunct to routine air pollution monitoring activities. While there are limitations in astronomical data, these are usually smaller and better known than the errors in other types of data. Astronomers can provide (i) nighttime extinction data, (ii) a new set of remote locations as well as some new urban ones, (iii) a lengthy data record (beginning in 1902 in the case of Abbot's data), and (iv) a variety of new measuring techniques.

Some conclusions reached through astronomical research that are of special interest to atmospheric scientists are as follows:

1) No global trends in extinction that can be ascribed to human causes have been detected at remote locations so far.

2) Both volcanic activity and some apparently natural cycling of the biosphere affect astronomical extinction; the latter effect produces an annual pattern.

3) The transparency of the atmosphere near cities continues to deteriorate in most cases.

4) The anomalous wavelength dependence of extinction suggests a unique aerosol size distribution in background locations.

Also from the meteorological viewpoint, a number of gaps became apparent in available knowledge. Perhaps the most obvious one is the need for coordinating extinction measurements with local and synoptic meteorological data. Certain biases exist because of the lack of extinction data for cloudy periods. Also, effects have been discovered that are due to dust derived from upslope winds on mountaintops. Since regional effects of human activity will certainly be apparent before global effects, studies of the transport of air masses to and from observing sites may be possible.

Finally, another kind of pollution is of less concern to meteorologists but of vital concern to astronomers. Light pollution, caused by the rapid expansion of outdoor lighting in cities and even in open western countryside, has been of great concern in many observatories recently. Greater public awareness of the problem, research on more efficient lamps (from the point of view of increased ground illumination and reduced light loss to the sky), and political action through the introduction of city ordinances are among the

goals of astronomers in various research centers of the Southwest.

At the Seattle meeting astronomers emphasized their concern about the deterioration of the atmosphere, through which most of the information about the universe must pass. Progress in the application of certain kinds of astronomical data to research on atmospheric deterioration has begun. Much more must be done to understand fully the implications of the data and even more, of course, to turn the tide. Charles Abbot may not have expected that his data and methods would be used for these purposes, but his are the best optical data available over such a long time base and they have come to us only because of his painstaking care in gathering and recording all available information in his experiments.

P. W. HODGE  
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### References

1. N. Laulainen and P. W. Hodge, Eds., *Project ASTRA Publication 15* (Univ. of Washington, Seattle, 1972).
2. C. G. Abbot, *The Sun and the Welfare of Man* (Smithsonian Institution, New York, 1929), p. 68.

### Forthcoming Events

#### January

9-12. American Astronomical Soc., Las Cruces, N.M. (H. M. Gurin, AAS, 211 FitzRandolph Rd., Princeton, N.J. 08540)

9-13. National Soc. of Professional Engineers, Salt Lake City, Utah. (P. H. Robbins, NSPE, 2029 K St., NW, Washington, D.C. 20006)

14-19. Protein Phosphorylation in Control Mechanisms, Miami, Fla. (W. J. Whelan, Dept. of Biochemistry, School of Medicine, Univ. of Miami, P.O. Box 875, Biscayne Annex, Miami 33152)

15-16. Regional Environmental Management Conf., San Diego, Calif. [L. E. Coate, REMC, County of San Diego, Environmental Development Agency, Integrated Regional Environmental Management (IREM) Project, 1600 Pacific Hwy., San Diego 92101]

15-17. Lunar Dynamics and Observational Coordinate Systems, Houston, Tex. (J. D. Mulholland, Lunar Science Inst., 3303 NASA Rd. 1, Houston 77058)

15-18. American Crystallographic Assoc., Gainesville, Fla. (Mrs. E. E. Snider, ACA, 335 E. 45 St., New York 10017)

15-19. Geophysics of the Earth and the Oceans, 2nd intern. conf., Australian Inst. of Physics and Australian Soc. of Exploration Geophysicists, Sydney. (B. D.